FEDERAL/PROVINCIAL
RESEARCH AND MONITORING
COORDINATING COMMITTEE (RMCC)

ASSESSMENT OF THE STATE OF KNOWLEDGE ON THE LONG-RANGE TRANSPORT OF AIR POLLUTANTS AND ACID DEPOSITION

PART 3

AQUATIC EFFECTS

AUGUST 1986

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PLUIES ACIDES

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RMCC AQUATIC EFFECTS SUBGROUP

AUGUST 1986

AUTHORSHIP OF THIS ASSESSMENT REPORT

A special Task Force was appointed under the Federal/Provincial Research and Monitoring Coordinating Committee of the Long Range Transport of Air Pollutants Program, to undertake assessment of the present status of information related to the acidification and its effects on the Aquatic Ecosystems in Canada. Members of the Task Force authored individual sections of the report and have contributed to review and editing of the entire report.

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INTRODUCTION

The Federal/Provincial LRTAP Research and Moitoring Coordinating Committee (RMCC) has been directed to undertake an assessment of the present status of information in relation to acidification of the various ecosystem components.

The report focuses on the Canadian aquatic ecosystem but makes use of other information as appropriate to establish a linkage on a specific effect. Information that has been published of research carried out since completion of the U.S.-Canada Memorandum of Intent (MOI) (Environment Canada, 1982) report are the primary sources. It is assumed that readers will have as a basic background the MOI Report on Impact Assessment, Workgroup I. (The acronym LRTAP is used in this report for Long Range Transport of Airborne Polluants).

FORMAT OF REPORT

The report is developed in terms of the degree to which the research and monitoring programs have provided evidence in relation to specific questions or issues concerning the acidification and its impact on the aquatic ecosystem. The questions consist of a single primary and several secondary questions that are addressed.

Primary Question

> What is the relationship between areal loadings of atmospherically deposited pollutants and the alteration of the chemical and biological status of freshwater aquatic ecosystems in Canada? Is the present policy "target loading" of 20 kg/ha.yr of wet sulphate protective of these ecosystems as defined by quantitative measure of susceptibility?

Secondary Question

1. What is the evidence that the chemical characteristics of aquatic receptors have changed significantly over the past several decades and what are the regional variations of the changes?
2. What is the evidence that changes in aquatic chemistry are caused by the deposition of atmospherically transported material?
3. What is the evidence for changes in the aquatic biological community during the same period?

4. What evidence is there of a relationship between the chemical changes and the biological status of the aquatic ecosystem?

5. Can the rate of response and rate of return to equilibruim under a new level of deposition stress be defined quantitatively?

6. What are the potential consequences of mitigative measures?

Final Question:

What portion of Canada's aquatic ecosystem resoures are at risk and what is the projected damage under specified deposition rates?

The assessment report depends upon research results that are available in either the open literature or in agency reports that can be obtained for review. The report will also draw upon information that was presented at the Muskoka International Symposium on Acid Precipitation, September, 1985. These papers are referred to as "in press" although the prepublication reviews are still in progress.

The assessment report is considered as an interim assessment in that the LRTAP research program will serve to identify areas where information is incomplete in relation to the issue questions and can serve to direct the research in the future program.

ASSESSMENT OF AQUATIC ECOSYSTEM ISSUES

The primary question can be addressed only after information has been examined and answers have been obtained to the secondary supporting questions. The questions are, therefore, examined in that order.

3.1 What is the evidence that the chemical characteristics of aquatic receptors have changed significantly over the past several decades and what are the regional variations of the changes?

3.1.1 Evidence of Changes in Surface Water Characteristics

Since the major analysis of existing information represented by the U.S.-Canada, Memorandum of Intent (Environment Canada, 1982), several studies have provided information which confirms and/or strengthens the conclusions regarding temporal and spatial changes in surface

water composition arising from the influence of LRTAP. No reevaluation of historical water quality data has been attempted since the MOI;, however new paleolimnological studies have provided substantial indirect evidence for the temporal alteration of remote lake water quality due to LRTAP. Nriagu and Coker (1983), Nriagu and Soon (1985) and Carignan (1984) have shown that variations in the content of sulphur (and its various forms) in lake sediments can be used to infer historical changes in the sulphur cycle of the lakes. In some lakes where surface perturbations did not occur (biological and/or physical), near surface increases in total S concentration (with concommitant Pb-210 dating) show that remote lakes in Ontario and Quebec probably experienced elevated lake water sulphate concentrations beginning around 1890. More dramatic increases in sediment concentrations for lakes near the large point source at Sudbury closely correspond to the known dates of initial smelting at this site, while slightly more distant lakes show the influence of the later installation of low level stacks. This information plus the large body of existing and new information on historical variations in metal sedimentation in lakes (Evans et al. (in press), Evans and Dillon (1982), Wong et al. 1984), Johnson (in press) shows that lakes in eastern Canada have experienced LRTAP related perturbations in their water quality for 80-100 years. The degree of change in water quality has been greater in recent times.

Several evaluations of past lake acidity using the fossil diatom technique (Charles, 1984; Charles et al. in press; Smol et al., 1984) suggest that significant decreases in pH occurred only during the last 30-40 (e.g. post-war) years and only for some lakes, presumably those with a major buffering limitation or those specifically affected by a nearby source. Areas which have had a longer industrial history such as the U.K. show earlier occurrences of major pH reduction (Battarbee et al. 1985). All of the above evidence confirms that surface waters have experienced higher sulphate concentrations, higher acidity (and therefore necessarily lower alkalinities), and higher metal (particularly Pb) levels over the past several decades. The quantitative magnitude of these effects vary spatially.

The regional variability in existing surface water quality is much better established now than at the time of the MOI due to:

1) continued extensive survey work using better lake selection, sampling and analytical methodologies than were used in prior surveys, and
2) the collation, computerization and evaluation of data from several different provincial, university and federal sources (true in both Canada and USA).

In a broad regional sense, Jeffries et al. (in press) have shown that the most sensitive waters in eastern Canada (as shown by low Ca + Mg concentrations, Table 1) occur in the Maritime provinces and that the variability in sulphate and acid neutralizing capacity (alkalinity)

Table 1. Median lakewater concentrations of Ca+Mg, alkalinity and sulphate for subregions of eastern Canada as defined by Jeffries _et al_. (in press)

Subregion	n*	Median Parameter Concentration(μeq/L)		
		Ca + Mg**	Alkalinity	SO_4**
Nova Scotia	275-413	53	10	50
New Brunswick	52-81	90	38	77
Newfoundland	176-268	79	28	31
Labrador	180-203	69	46	24
Quebec	246-429	141	36	82
South-central Ontario	750-1578	199	63	158
North-eastern Ontario	793-1805	441	165	148
North-western Ontario	216-1078	221	254	75

* Number of data used to determine median varies among parameters.
**Sea-salt corrected.

across regions can be explained by a combination of deposition and geochemical (terrain) factors. Similar conclusions were reached by Kelso _et al_. (1986). A high percentage of acid lakes occurs in the _extremely_ sensitive terrain of southern Nova Scotia that receives wet sulphate deposition very near the present target loading of 20 kg/ha/yr. This suggests that the target may be too high for the protection of these highly sensitive waters; however, it should be noted that the target loading was proposed "to protect all but the most sensitive aquatic ecosystems in Canada".

It must be emphasized that this is a static assessment, i.e., presenting the status over a short time period, approximately the last five years. In order to assess the magnitude of water quality change over time (but not the rate of change), it is necessary to look at the relationships _among_ major ion concentrations using the assumption that the water quality prior to LRTAP influences was similar to that of current waters found in similar terrain but which receive low deposition levels. This approach has been applied by Wright (1983) to 27 data sets in Europe and North America. The empirical model or "acidification equation" determined by Wright shows that an increase in sulphate deposition must result in an increase in acidity (equivalent to a decrease in alkalinity) or an increase in the base cation concentration or in an increase partitioned between acidity and base cations. Several studies, for example Thompson (in press), suggest that the former effect predominates although Wright does point out that the latter may occur in the short term as the readily exchangeable base cation content of soils

is leached during early acidification stages. From data collected in low deposition areas, Wright (1983) also suggests that the "normal" sulphate level expected to occur in lakes is in the range 26-41 ueq/L. Similarly, Talbot et al. (1984) suggest a value of 30 ueq/L for the remote northeastern part of Quebec. All of these values are substantially less than the median levels reported by Jeffries et al. (in press) for NW ONT (73 ueq/L), NE ONT (138), SC ONT (159), QUE (82), NB (82) and NS (56). It should be noted again that this type of analysis does not say anything about the rate of change, only about the type of changes in water quality.

3.1.2 Evidence of Changes in Groundwater Chemistry

In Canada, evidence for strong mineral acid weathering has been obtained from studies conducted in the Turkey Lakes Watershed, Sault Ste. Marie, Ontario (Craig et al. 1986 and Johnston et al. 1985). In the near surface (1 m), groundwater exhibited a pH depression from 5.2 to 4.6 and an alkalinity decrease from 25 to 0 μeq/L in response to H^+ loading events. At intermediate depths (1-3 m), the absolute changes in pH and alkalinity were similar but the initial values are much higher (5.7 and 6.6, and 60 and 400 μeq/L). These variations are related to the input of SO_4 and NO_3 from the melting snow pack.

Monthly sampling data from Chalk River, Ontario indicate a pH decrease of 0.5 pH units and a decrease in alkalinity from 160 to 80 μeq/L over a three year period (Johnston et al., 1986; Flavelle, 1985). There is a concurrent increase in sulphate from 200 to 400 μeq/L. The interpretation of the Chalk River results is complicated by the presence of road salt contamination in the area.

A study of the groundwater regime at Lac Laflamme, Quebec (Azzaria and Gelinas, 1981) documented the presence of acidic groundwater (pH 4.5 to 4.4) in shallow (<1.4 m) zones of rapid infiltration. Concurrent with low pH values the samples contained lower concentrations of alkalinity, specific conductance, Ca and Mg. At greater depths, groundwater pH > 6.0 was common. The sampling period is too restricted to allow conclusions concerning the long term to be drawn.

Table 2. Groundwater Chemistry, Lac Laflamme, Quebec Azzaria and Gelinas (1981).

Depth	pH	Alk (mg/L $CaCO_3$)
1.12	4.05 - 5.61	5.3 - 6.0
1.21	4.30 - 5.85	0.5 - 8.0
1.34	4.95 - 6.35	4.0 - 9.0

In a shallow sand aquifer near Pinawa, Manitoba, Robertson and Cherry (in press) report a 20-fold increase in $SO^{=}_4$ levels in groundwater less than 30 years old, compared to groundwaters which are 60 to 240 years old. These increased levels start to appear in groundwater recharged slightly before 1950 and continue in most recently recharged groundwaters. These waters are high in bicarbonate (pH 6.5 to 7.0) and no pH depression has been noted.

A one time sampling of groundwater near Sudbury, Ontario (Sibul and Reynolds, 1982) indicated that 5% of 125 sites had pH < 6.0 but neither the long-term trends nor the seasonal trends were considered. A similar survey in the Muskoka-Haliburton area gave similar results (Sibul and Vallery, 1982).

In Europe, documented evidence for groundwater acidification in non-calcareous terrain is scarce but there is a growing body of circumstantial evidence that points to acidification of shallow groundwaters. Hultberg and Johannsson (1981) and Jacks and Maxe (1984) report acidified groundwater from Sweden, especially in shallow wells. Grimvall et al. (1985), in their discussion of quality trends in public water supplies in Sweden, state that a "national trend of groundwater acidification is in fact statistically verified" based on a drop in the alkalinity to hardness ratio. Groundwater acidification was identified as a major problem in the Federal Republic of Germany (Schoen et al. in press; Wittig in press). Changes in groundwater pH were serious enough to result in the closure of several small municipal water supplies. Acidification of shallow groundwater was also reported by Krieter (1985) and Puhe (1982).

Tamm and Hallbacken (in press) reported a change in soil pH of 0.5 to 0.7 units under different tree canopies in southwest Sweden resampled at a 50-year interval. Varying degrees of soil acidification were reported for three Danish spruce forest ecosystems (Rasmussen in press).

3.1.2.1 Link between Groundwater and Surface Water Chemistry

A question which has received less attention concerns the influence of groundwater chemistry on surface water chemistry. Bottomley et al. (1984) concluded that for the two areas studied that groundwater had higher alkalinity than the surface water and that "groundwater discharge results in significant neutralization of runoff acidity". Where sufficient overburden exists groundwater contributes 50% of the water in headwater streams during springmelt and approximately 70% during summer storms. During summer many streams would cease to flow without groundwater.

Henriksen et al. (1984) discussed the transport of Al-rich groundwater to surface waters where the subsequent CO_2 degassing resulted in removal of HCO_3, hydrolyzation of Al and precipitation of Al. In rivers, the precipitate will be carried downstream and parts of it will be absorbed to the surface of the substrate. In rivers with large pH fluctuations, episodes with low pH will dissolve precipitated aluminum and create conditions that will be toxic to fish.

Prenzel (1983) had postulated the formation of a new compound ($AlOHSO_4$) when there is $SO^=_4$ weathering of aluminum silicates. This compound will act as a sulphate-sink for some years but further decreases in pH result in its disintegration and a rising output of $SO^=_4$ from the subsurface system. Similar results were obtained by Hauhs (1984) who further suggests that a "break through" process may occur when the storage capacity is exceeded and acidification increases.

Furthermore, some fish species instinctively search out groundwater for use as spawning areas. They rely on the consistency and quality of groundwater chemistry and the benefits that can be accrued even in surface water systems that are suffering from acidification.

In summary, groundwater is less acidic and has much higher alkalinity concentrations than co-existing surface water. If, at some future date, groundwater becomes more acidic and substantial decreases in alkalinity result, the groundwater system will no longer be able to improve the chemical quality of the surface water system. The resulting rapid degradation in quality will greatly diminish its ability to support life.

3.1.3 **Models of Chemical Changes**

(a) Observed improvement in pH conditions in surface waters due to loading decrease near Sudbury and in Nova Scotia rivers has been confirmed by model results.
(b) Observed pH depressions due to episodic snowmelt events can be accurately simulated by models.
(c) Lake chemistry changes and their linkage to SO_4= deposition and sediment reduction have been incorporated in simulation models only recently; more work needs to be done.
(d) Groundwater chemistry changes are most difficult to model, because of lack of data on reliable weathering rates.

Chemical changes in the aquatic regimes can be detected or confirmed by applying mathematical models against observed data. In the case of surface waters, Thompson (in press) shows that the observed increase in pH in Nova Scotia rivers over a time span of a decade can be attributable to the decrease in excess sulphate over the same period.

Dillon and Girard (in press); and Johnson et al. (1985) also found rapid recovery in the pH (about one pH unit rise over a time span of five years) in the Sudbury area where sulphur emissions were reduced by 80% from the early 1970's to the early 1980's. However, when the Thompson-Henriksen CDR model is applied, a correction factor, F=0.75, representing the change in base cation concentration per unit change in sulphate concentration, is required. The necessity of introducing the so-called F-factor is also reported by Wright and Henrikson (1983), Fraser (in press) and Minns and Kelso (in press). The values of the F-factor vary between 0.2 to 0.8 and are dependent upon the sulphate adsorption rate, weathering rate, soil contact time. The inclusion of nitric and organic acids may also be necessary, in some cases.

More in-depth investigation of the relationships between the terrestrial characteristics and the changes in the aquatic regimes has lead to the development of several process-oriented models. A recent review of these models has been given by Christophersen and Seip (in press), Reuss et al. (1985) and Johnson et al. (1985). Typically, these models consider the soil compartment as an ion-exchange system in which chemical ions can be exchanged rapidly so that chemical equilibrium relationships are applicable. The cation exchange capacity is assumed to be much larger than the amount in the solution, but the base saturation, i.e. the fraction of adsorbed base cations, can be reduced over time. Thus, the time-dependent changes in both the water chemistry and soil chemistry are linked, in contrast to the steady-state assumptions of the CDR models. The tradeoff is that more model coefficients are introduced and have to be related to experimental data (e.g. Dupont and Grimard in press).

Therefore, the calibration and verification of these process-oriented models depend on the results of very comprehensive watershed experiments in both the terrestrial and aquatic regimes. So far, these models have been successful where a comprehensive set of watershed data are available. For example, the Christophersen and Seip Model has been applied to several watersheds, e.g. Birkenes and Storgama in Norway (in press), Turkey Lakes, Lam et al. (in press) and Harp Lake, Rustad et al. (in press) in Canada and Plynlimon in Scotland, Whitehead et al. (in press). In each case, the observed changes in the water chemistry are short-term, seasonal or episodic events and the changes, particularly pH drops due to snowmelt, are accurately reproduced by the model if the hydrology is accurately simulated (Bobba et al. in press; Scheider and Logan in press; Rustad et al. in press; Morris and Thomas in press; Jones et al. in press; Goodison et al in press). Likewise, hydrology and the terrestial characteristics (April et al. 1985) play a key role in the simulation of the water chemistry in Adirondack lakes using the ILWAS model (Chen et al. 1983. Thus, these process-oriented models have sufficiently advanced to simulate water chemistry changes on a seasonal or annual time scale.

In the case of groundwater modelling, the availability of data is the limiting factor. Observations such as those conducted by Johnston et al. (1985) have been used to define the water pathways in Turkey Lakes, Bobba et al. (in press) and Harp Lake, Rustad et al. (in press) by the ^{18}O tracer data. Increase of groundwater input of calcium in the downstream direction has been simulated (Lam et al. in press) and has been used to explain the observed spatial gradient of pH, alkalinity and even algal primary production in Turkey Lakes (Lam et al. 1985). However, the long-term simulation of the chemical changes in groundwater has been largely limited to theoretical investigation (e.g. Galloway et al. 1983; Cosby et al. 1985). Simulation results of the groundwater chemistry over decades and centuries are difficult to verify, although some success has been met by using reconstructed paleolimnological data (Wright et al. in press) using the MAGIC model (Model of Acidification in Groundwater in Catchments). This model focuses on detailed exchanges between major ions in the groundwater particularly $SO_4^=$, Al^{3+} and HCO_3^-. A number of hypotheses used in the model, such as the dependency on soil temperature in the formulation of CO_2 degassing, still need to be confirmed.

New experimental results on the sulphate reduction as a source of alkalinity generation at the lake sediments (Schindler et al. in press; Schiff and Anderson in press; Dillon et al. in press) have implications on sulphate budget and the sulphur loading reduction program. For example, the Trickle-Down Model by Schnoor et al. (1982), which is based on the concept of acid neutralizing capacity (ANC), i.e. alkalinity, has been extended to investigate the ANC generation in seepage lakes. Model results for Lake Vandercook show 24% of alkalinity originated from groundwater and 76% from sediments and other internal lake processes (Lin and Schnoor 1985). However, the actual rate of ANC production is very small, in the order of 10^{-4} to 10^{-6} day^{-1}, and the groundwater residence time is long (150 years). Thus, it is expected that these proportions may change substantially in non-seepage lakes. A more detailed model on the same subject is designed by Baker et al. (in press) which include sulphate reduction, nitrogen assimilation, cation exchange and weathering. However, the kinetics are all first order and are based on lake data from Florida, Wisconsin and Minnesota where acid deposition is relatively mild. It is not known whether these rates (e.g. sulphate reduction rate, K_{SO_4} = 56.5 meq/m^3yr) would hold in other lakes.

3.2 What is the evidence that changes in aquatic chemistry are caused by the deposition of atmospherically transported material?

3.2.1 Evidence of Direct Relationships and Regional Comparison

The most important ions in deposition are hydrogen ion (or negative alkalinity), sulphate, nitrate and ammonium. Of these, sulphate is

the only parameter which often exhibits an approximate balance between inputs and outputs for lakes in eastern Canada. Therefore it is not surprising that sulphate exhibits the best relationship between deposition and lake concentrations. For example, Jeffries et al. (in press) show that the median regional sulphate concentration in eastern Canada follows the expected gradient in deposition in support of earlier observations made by Talbot et al. (1984), Thompson and Hutton (1985) and Kelso et al. (1976). Similarly, Dupont and Grimard (in press) were able to show that a relationship exists between lake pH and precipitation sulphate concentration within specified sensitivity classes for a survey of 1091 lakes in Quebec. Regional studies in the USA have found also a simple, direct relationship between the intensity of sulphate deposition and lake concentrations [Hendry (in press) for New York and New England and Rapp et al. (1985) and Glass and Loucks (1986) for the northern Great Lakes states]. Failure to observe a simple relationship can usually be attributed to either sulphate adsorption in the terrestrial basin (commonplace in southern, unglaciated regions of the USA), or to large local variations in deposition caused by local sources or topographic control of precipitation quantity. The implication of this second point is that if a more precise characterization of local deposition were available than is now possible, these "outliers" would probably fit the simple relationship.

Alkalinity concentrations show a poorly defined relationship with atmospheric deposition due to the many confounding factors associated with its input to lakes, e.g. influence of bedrock and surficial geology, forest type, other terrain characteristics such as the presence of wetlands, within-lake generation, etc.

Significant within-lake sources of alkalinity include permanent sulphate and nitrate reduction, and co-diffusion with calcium (or other like ion) from the sediments. Carignan (1985) and Schindler (in press) both consider that such within-lake processes are significant to the alkalinity budget of lakes. Such processes are variable from lake to lake (Schiff and Anderson in press) and in some cases may be difficult to distinguish from alkalinity entering the lake via groundwater seepage (Jeffries et al. in press). In few cases, the groundwater may influence surface water chemistry to the extent that the influence of atmospheric deposition is masked.

Throughout most of eastern Canada and the USA, atmospherically deposited nitrogen species (ammonium and nitrate) are utilized as nutrients within the terrestrial or aquatic ecosystems. Hence, there is no discernable relationship between deposition and surface water concentrations for these two parameters. However, there is some indication that the nitrogen nutrition requirement has reached or is reaching saturation for some locations (Likens et al. 1977; Cronan, 1985; Jeffries et al. in press) and thus a defineable relationship may emerge in the future. The decreased need for or retention of nitrate

by the terrestrial ecosystem may have important implications with respect to aluminum biogeochemistry; for example, Driscoll (in press) reported that nitrate had a profound effect on the transport of aluminum to acidic waters. At present it is not known whether this observation is site specific.

The occurrence of acidic lakes has been linked to the presence of naturally occurring organic acids; indeed there is a well defined class of coloured (e.g. dystrophic) lakes which contain significant levels of organic acidity and are usually associated with wetland drainage. Several studies have been conducted to determine whether this mechanism of natural acidification can explain the widespread occurrence of clear acidic lakes. LaZerte and Dillon (1984) studied the relative contribution of natural organic and anthropogenic strong acids to the acidity of surface waters in Muskoka-Haliburton, Ontario and concluded that the strong acids are responsible for the acidification occurring there. Kerekes et al. (in press) similarly studied the acidity components of highly organic Nova Scotia waters. They concluded that while the presence of organics in streamwaters may result in a 1 unit lower pH than that in equivalent clear water, observed pH declines well beyond this level were due to the presence of anthropogenic sulphate. Finally, Jeffries et al. (in press) presented statistics showing the distribution of lake alkalinity within classes of increasing organic anion concentrations; the even distribution of alkalinity across all classes of organic anion concentration means that lake acidification by naturally occurring organic acids is not a predominating occurrence in eastern Canada. Davis et al. (1985) have shown that acidification by strong acid inputs causes a decrease in the organic carbon content of lake water. Since highly coloured (i.e. organic rich) lakes have a substantially different biogeochemistry than clear lakes (Such as Al and other metal complexation), loss of organic carbon can be expected to cause major changes in the overall ecology of these lakes.

3.2.2 Models of Linkages of Atmospheric Deposition and Aquatic Chemistry

Much of the work on watershed modelling has been devoted to short-term episodes covering several years. The linkages to atmospheric deposition are confined to such episodes as snowmelt events, with watershed model results. On the long-term basis, the CDR model results (Thompson in press) consolidate the linkage to atmospheric loading with data from Nova Scotia rivers (Clair in press). Dependence of sulphate in surface waters on the atmospheric loading is generally recognized. It is less clear how the alkalinity and pH may respond for given watershed and lake characteristics. The influence of precipitation sulphate is strong in the analysis of diatom data using the MAGIC Model (Wright et al. in press), but the lake sulphate concentration is also a function of weathering rate, sulphate

adsorption, lime potential and percentage base saturation. In general, if these functions are assumed to be known, direct linkage to atmospheric deposition can be established by process-oriented models, particularly those using the mobile ion concept. For example, predictions of lower sulphate concentration and higher pH can be made for lower loading at the Harp Lake Watershed, even though the model has only been calibrated with short-term data (Rustad et al. in press; Christopersen et al. in press).

It is also clear that model predictions are associated with uncertainties which are non-trivial subjects themselves (Cosby et al. 1985). The uncertainties could be due to measurement input errors, model coefficients, model formulations and stochastic noises, etc., for example, in Thompson (in press), the prediction of the stream pH for the Nova Scotia paper is well within 0.2 or 0.3 unit of pH. Translated to the uncertainty of the estimated CDR, it could mean about 20% error margin in the CDR. It could mean other types of uncertainties as well. Thus, uncertainty analysis is definitely required when the water chemistry is linked to the atmospheric loading.

3.3 What is the evidence for changes in the aquatic biological community during the same time periods?

3.3.1 What Direct Evidence is There of Change in Fisheries?

In areas removed from point source influences, historical evidence of change in fisheries exists.

In Canada, several examples of change in fisheries communities have been documented by the U.S.-Canada, Memorandum of Intent (Environment Canada, 1982) and Harvey (1982). In Northeastern Ontario (Algoma area), Kerr (1982) and Beggs et al. (1985a) found evidence of fish population losses (particularly lake trout) in acidic lakes located beyond the point source influence of the Wawa smelters. Kelso and Gray (1984) also found that, in the Algoma region, lakes existed where a change in chemistry coincided with population declines and losses. However, as in most comparisons with historical information, chemical methodologies differed through time and the fisheries data were not always consistent or reliable.

The Adirondack region in the United States is one of the largest sensitive lake districts in the eastern U.S. Pfeiffer and Festa (1980) suggest that the greatest negative impact has been exerted on the brook trout fishery. Further, in a survey of 46 states, 20% (nine states) asserted that atmospheric precipitation is currently exerting a negative impact on the state's fishery resources (Pfeiffer, 1982). The nine states claiming a negative impact upon their fisheries indicated that 4100 ha of surface waters have been acidified. Haines

and Baker (in press) in an examination of historical data from nine states reported that the strongest evidence supporting the hypothesis that acidification reduced or eliminated fish populations in the eastern United States came from the Adirondack region of New York. There, a high probability exists that fish communities have declined as a result of acidification in 49 lakes. Weaker evidence exists for declines in fish populations in Pennsylvania and Massachusetts.

One of the most complete and convincing case histories is the record of decline in angling catch of Atlantic salmon from Nova Scotia rivers of pH < 5.0 (Watt et al. 1983; Watt in press). These records have been corroborated with evidence of the effect of low pH on Atlantic salmon in laboratory experiments (Daye, 1980; Peterson and Martin-Robichaud 1983; Peterson 1984, etc.) in in situ bioassays (Lacroix 1985; Lacroix et al. 1985), and electrofishing surveys (Watt et al. 1983).

In two interesting cases near Sudbury, low pH had reduced the lake trout population in Whitepine Lake to a relict population of a few large adults (Gunn and Keller 1984a; Beggs et al. 1985b) and the brook trout population in Joe Lake had been extirpated (Beggs et al. in press). For both of these populations, recruitment resumed (brook trout were stocked in Joe Lake) with the concomitant and natural improvements in water quality likely due to reduction in smelter emissions in the Sudbury area (Beggs et al. in press). These cases exemplify the linkage between population success and water quality and also enhance the cases for continued efforts to improve habitat quality.

In Scandinavia, where a good long-term historical record exists, losses of fish stocks are still occurring (Sevaldrud and Skogheim in press). Although the decline in fisheries was most severe during the 1960's and 1970's, population losses are still occurring. Between 1978 and 1983, brown trout and yellow perch populations were still being extirpated concomitant with a deterioration in water quality.

3.3.1.1 What Direct Evidence is there of Stress to Fisheries?

Direct evidence of stress in fish exposed to acidified natural waters has been repeatedly demonstrated using in situ bioassays (selected examples in Table 3). In these exposures, the mortality of wild and hatchery-reared fish of many species has been linked primarily to elevated concentrations of hydrogen ion. In some cases, high concentrations of aluminum have also been implicated, however, the identification of the causative agent(s) is clouded by the lack of rigorous control that is possible in laboratory bioassays. The clearest examples of mortality induced by acidity come from the results of exposures compared with limestone-altered reference exposures. For example, mortality of lake trout and brook trout

TABLE 3. A summary of _in situ_ bioassay results

Species	Life Stage	Results	Reference
Lake trout and brook trout	Eyed eggs - emergence	Alevin emergence improved in limestone (pH>6) over gravel (pH 5.5) treatment	Gunn and Keller 1981
	Eyed eggs - sac fry	Mortality of sac fry (18%) coincided with maximum surface runoff and peak concentrations of inorganic aluminum. (Incubated above natural spawning shoal).	Gunn and Keller 1984
	Emergence-exogenous feeding.	<1% mortality.	
Brook trout (Aurora trout)	Fry	Mortality at pH <4.8 significantly greater than mortality at reference site (pH 6.5).	Beggs _et al._ 1985
Smallmouth bass	Fry	Mortality at pH < 5.15 significantly higher than mortality at a reference site (pH 6.0).	
Walleye	Fertilized eggs-hatch	Mortality to hatch high at pH 5.4 relative to a reference at pH 6.6.	Hulsman and Gunn 1983
Rainbow trout	Yolk sac fry	100% mortality at sites of pH 5.4 and 4.6 as compared with 1.3% mortality at reference site (pH 6.6).	Hulsman and Gunn 1983
Lake trout Brook trout	Alevin and yearling	Exposed in littoral zone of Plastic Lake - survival negatively correlated with hydrogen ion concentration.	Munro _et al._ (in press)
Atlantic salmon	Eggs - embryos	Hatching success in natural substrate of acidic rivers is positively correlated with pH of interstitial water. Emergence of fry reduced at pH < 4.8	Lacroix 1985
	Emergence fry	In water of pH 5.0 mortality of post-emergent fry range was 70% possibly due to the inability to initiate exogenous feeding. Mortality in limestone treated water (pH 6.1) was 4%.	Lacroix _et al._ 1985
	Adult	Metabolism of androgenic and steroid hormones of caged fish abnormal in an acidified river (pH 4.7). Fish also had less weight gain, produced smaller eggs, and eggs were less viable than fish held in a reference river (5.6 pH).	Freeman and Sangalang 1985

alevins incubated in containers of gravel (pH 5.5) was significantly higher than mortality of these species incubated in limestone substrates (pH > 6.0) (Gunn and Keller 1981). Similarly, Lacroix et al. (1985) showed that mortality of post-emergent Atlantic salmon fry in limestone treated water (pH 6.1) was significantly lower than mortality of fry in untreated water (pH 5.0).

While the results of the in situ bioassays (summarized in Table 3) clearly demonstrate the loss of potential fish habitat due to low pH, extrapolation of the results to effects on natural populations is confounded by several factors:

fish are constrained by exposure containers and hence are not afforded the opportunity to seek refuge and avoid lethal conditions. Laboratory studies using a variety of fish species have shown that often fish avoid acidic water at sublethal pH levels (Van Collie et al. 1983; Jones et al. 1985; reviewed in Gunn in press). Even the earliest mobile life stage of brook trout, the emergent fry, has been shown to avoid acidic water (Gunn and Noakes in press). Low pH in combination with high concentrations of aluminum was also actively avoided by juvenile Atlantic salmon (Van Collie et al. 1983). However, the results of these laboratory avoidance experiments remain largely unconfirmed in the field. Hall et al. (1980) observed the movement of adult brook trout downstream from a section of stream that was experimentally acidified and avoidance has been implied by observations of fish distribution patterns (Muniz and Lievestad, 1980; Watt et al. 1983).

3.3.2 Indirect or Empirical Evidence of Linkage between Aquatic Chemistry and Fish Population Status

3.3.2.1 Empirical Relations through Surveys

Although most surveys were designed to either form a basis for estimating the resources at risk or establishing the current status of the resources, the data bases have often been used to examine the relation between fish community structure or biomass (usually as reflected by catch-per-unit-effort) and lake chemical status. These empirically derived relations, although not directly linking cause (acidification) with effect (fish population losses) do circumstantially link the two. Collectively, these surveys have correlated lower fish species diversity with low pH, lower catch/effort with lower pH's and species presence/absence with ranges in pH. Sources of these observations, references and summary results are listed in Table 4.

Table 4. Surveys or Studies that Relate Fish Community Structure or Biomass to Water Chemistry.

Reference	Result
Smith _et al_. 1985	Species diversity and richness in relation to chemistry for 234 Nova Scotia lakes.
Rahel & Magnuson in press	Species occurrences linked with chemical variables for 31 species in 138 Wisconsin lakes.
Richard _et al_. in press	Linked density of brook trout with chemical characteristics and species diversity with chemistry for Quebec lakes.
Matuszek in press	For 3000 Ontario lakes showed the limiting effect of pH on species number independent of the limiting effect of lake size.
Wales in press	For 3000 Ontario lakes showed the limited occurrence of many species at low pH.
Pauwels in press	in 22 Maine lakes, number of fish species significantly related to pH.
Kelso _et al_. in press	Species diversity and abundance (CUE) linked with lake chemistry/morphometry for 832 east Canadian lakes.
Beggs _et al_. in press	Lake trout recruitment and abundance (CUE) and abundance of all species reduced a low pH.
Beggs _et al_. 1985	Brook trout abundance (CUE) reduced at low pH.
Weiner _et al_. 1984	Species richness lower in six acidic lakes as compared with six circumneutral lakes.
Kelso and Gunn 1984	Abundance (CUE) linked with lake morphometry and pH.
Canfield _et al_. 1985	Growth and condition of large mouth bass suppressed in acidic lakes. Plasna osmolalities lower in fish from clear, acidic lakes.
Frenette _et al_. 1984	Abundance (CUE) decreased with increasing acidity, condition factor linked to pH.
Lacroix _et al_. 1985	Atlantic salmon parrdensity lower in rivers of lower pH.
Bendell and McNicol in press	Presence of six cyprinid species in Ontario headwater lakes linked to pH.
Lacroix 1985	Showed differences in physiological condition of Atlantic salmon in rivers of pH < 5.0.
Freeman _et al_. 1985	Showed lower hormone levels in Atlantic salmon from acidified river.
Jagoe and Haines 1986	Found a few paired meristic and morphometric characters more asymmetric in brook trout and white suckers from low pH lakes.

Throughout the studies listed in Table 4, it is clear that the trend of fewer species and decreased diversity with increased acidity exists (Richard et al. 1985; Matuszek in press; Pauwels and Haines 1985; Weiner et al. 1984; Canfield 1985). The decrease in species richness begins when lake acidity increases to pH's less than 6. Also, many studies confirm that catch-per-unit-effort (reflecting abundance and/or density) declines with increasing acidity (Richard et al. 1985; Frenette et al. 1985; Kelso et al. 1986). Some studies (Frenette et al. 1985; Canfield et al. 1985; Ryan and Harvey 1980) have found that fish existing at lower pH levels either had reduced growth rates or lower condition factors; however, this response of fishes at lower pH is not always present (Kelso et al. 1986).

3.3.3 What is the Evidence of Effects of Acidification on Non-Fish Aquatic Biota?

There is increasing evidence that a wide range of non-fish aquatic organisms are very sensitive to low pH, and will not be protected by a critical criteria of pH 5.3. A variety of field and laboratory studies have demonstrated that several species of crustaceans and molluscs disappear when the pH is between 6.0 and 5.5. These include the three most common species of crayfish, the only freshwater shrimp in eastern North America, the most commonly abundant (in terms of biomass) species of zooplankton, *Daphnia galeata mendotae*, the most common and abundant amphipods of eastern North America, *Hyalella azteca*, and the most common and abundant snail of softwater lakes, *Amnicola limnosa*. Also there are several undesireable changes in the algal community at pH < 6.0. Several species of filamentous algae form massive, objectionable blooms in the littoral zone, and one planktonic species produces lakewide obnoxious odours. It is clear from these results that the present habitat criteria of pH 5.3 from which the target loading of 20 Kg SO_4/ha yr was derived will permit numerous adverse effects on aquatic ecosystems and that a much higher target pH (e.g. 5.8) is required for protection of these sensitive species.

Of the crustaceans, the sensitivity of three species of crayfish, *Orconectes propinquus*, *O. rusticus* and *O. virilis* is best established. Berrill et al. (1985) performed laboratory and transplant experiments with females of the first two species that were carrying broods of eggs or newly hatched young. At their control site, a hard water river at pH 7.5, all but one of 40 broods hatched and molted successfully through the first three larval stages, whereas at a stream site at pH 5.6 to 6.1 only 14 of 25 broods successfully reached the third stage. At two low pH sites (4.4-4.8 and 4.1-4.3) all broods failed and heavy mortality of the adult females was observed. In the laboratory, the success of the first two stages was assessed, and at pH 6.0-6.3 all ten of ten broods successfully molted, whereas at pH 5.4-5.9 only five of ten broods reached stage II, and

four of those five died before the end of the experiment on the eighth day. At pH 4.2-4.7 all broods and females died. These authors also surveyed ten lakes in the Killarney-Sudbury region and found O. propinquus only in the two lakes with the highest alkalinity (> 10 μeq/L).

The sensitivity of Orconectes virilis appears to be very similar to the other Orconectes species. During acidification of lake 223, no recruitment was observed below 5.63 (Schindler et al. 1985, Davies 1986), and a variety of other stresses on the adults were noted: calcium uptake was inhibited, hardening of the exoskelton was delayed (Malley 1980), parasitism by Thelohania greatly increased (France and Graham 1985), and egg mortality increased (France 1985).

The ecological significance of the loss of Orconectes species has not been assessed. It is known, however, that crayfish are the dominant food (60-90%) by volume of adult smallmouth bass and a significant component (10-40%) of the diet of largemouth bass (Scott and Crossman 1979). The loss of crayfish would force enhanced consumption of alternate prey, predominantly small fishes, unless the more tolerant but predominantly stream species of crayfish (Cambarus robustus and C. bartoni) replaced Orconectes.

The largest and most common species of zooplankton of glacial lakes (Roff et al. 1981), the freshwater shrimp, Mysis relicta, was eliminated during acidification of Lake 223 at pH 5.6 to 5.9 (Nero and Schindler 1983). The mechanism by which Mysis was affected was not rigorously established, but several possibilities (food availability, metal toxicity, increased predation, increased light intensity) were eliminated, and direct toxicity of hydrogen ion was suggested as the most plausible explanation. Nero and Schindler (1983) also cited examples of unsuccessful attempts to introduce Mysis in Swedish lakes pH values between 5.5 and 6.1. Stocking success was achieved only in lakes with values consistently above pH 6.0.

The ecological significance of the loss of Mysis due to lake acidification has not been directly studied, but a number of consequences can be inferred from changes that have been observed after the introduction of Mysis and from comparative analysis of plankton communities in mysid and non-mysid lakes. Mysis is a predator on zooplankton, and both native and introduced populations are associated with the absence or low abundance of certain species of Daphnia (especially hypolimnetic forms), Bosmina longirostris, Eubosmina spp., Cyclops scutifer and Diaptomus saugineas (Nero and Sprules 1986; Morgan et al. 1981). The loss of Mysis might allow abundances of these species to increase. A tremendous increase in Bosmina was observed after acidification of Lake 223. The net result in terms of prey availability for fish is difficult to predict, but

certainly the size distribution would shift towards smaller species of zooplankton that are more difficult to catch. Kinsten and Olsen (1981) observed an increase in phytoplankton in two lakes following introduction of Mysis and subsequent reduction of abundance of cladoceran grazers. A secondary effect of the loss of Mysis might be a decrease in phytoplankton, particularly the small edible forms.

Daphnia galeata mendotae is one of the most common cladoceran species of zooplankton in Precambrian Shield lakes of Ontario (Keller 1981; Roff et al. 1981), and in terms of biomass it often represents 25% of the annual average zooplankton standing crop (Hitchin and Yan 1983). Abundance of this species started to decline at about pH 5.6 during the acidification of Lake 223 and was eliminated the following year at pH 5.0. Consistent with this result is the low frequency of occurrence of D. galeata mendotae in lakes below pH 5.5 (Keller 1981). The mechanism by which this species is affected by low pH has not been elucidated, but data reported by Schindler et al. (1985) indicate no significant change in its food supply, and no increase in planktivorous predators in Lake 223. As Nero and Schindler (1983) discussed with respect to Mysis, metal ion toxicity was unlikely in Lake 223 and, by elimination, direct hydrogen ion toxicity is the most plausible explanation.

The ecological significance of the loss of Daphnia galeata mendotae has not been studied. It is well established that the large size classes of this species form a significant part of the diet of many fishes, but it is likely that other species will replace D. galeata mendotae when it disappears. For example, in Lake 223 D. galeata mendotae was replaced by D. catawba, a member of the relatively acid tolerant pulex group. (Schindler et al. 1985; Dodson 1981; Walton et al. 1982). In Muskoka-Haliburton the replacement species would probably be Holopedium gibberum. D. galeata mendotae and H. gibberum almost always co-occur in this area, but usually only one dominates the zooplankton (Hitchin and Yan 1983). Recent studies have focused on the grazing abilities of these and other species to explain their patterns of coexistence. It is well established from morphological analysis of filtering combs (Geller and Muller 1981) and laboratory studies of the grazing of sized particles (Hessen 1985) that D. galeata mendotae can filter even the smallest bacteria whereas H. gibberum can filter only the relatively large algae. Thus in situations where bacterial secondary production was significant, the replacement of D. galeata mendotae by H. gibberum would probably result in a decreased trophic transfer efficiency and decreased abundance of prey of the preferred size for fish. It should be emphasized, however, that the quantitative significance of these inferences is unclear.

The snail Amnicola limnosa occurs frequently with high abundance in non-acidified softwater lakes (Rooke and Mackie 1984a; Servos 1983; McKillop and Harrison 1972). Although the adults of this species are tolerant of low pH, Servos et al. (1985) have determined with laboratory bioassays that embryo development is delayed at pH 5.5 and totally arrested at 5.0 and below. In a quantitative survey of six low alkalinity (5 to 63 μeq/L) Ontario lakes, a significant decline in abundance with decreasing alkalinity was observed (Rooke and Mackie 1984). In one of the lakes (Heney) apparent reproductive failure was observed (Rook and Mackie 1984a, 1984b). Given the laboratory results on the sensitivity of embryonic development and the pH range of the lake (5.2 to 6.0) direct hydrogen ion stress on embryonic development was the most likely cause.

The ecological significance of the loss of Amnicola has not been investigated. Snails are known, however, to form a significant part of the diet of many fishes (Scott and Crossman 1979), and Amnicola is usually the most abundant snail in softwater lakes and streams. For example, in stream and lake surveys Amnicola comprised 33% and 61% respectively of all snails collected (McKillop and Harrison 1972; Rooke and Mackie 1984a). These observations suggest that the loss of Amnicola will force a significant change in the composition of the diet of many fishes.

The most common and abundant amphipod in eastern North America is Hyalella azteca (Pennak 1975). In a recent survey of Ontario lakes, Hyalella was found in 69 of 70 non-acidified lakes (alkalinity > 0) and in none of nine lakes that are presently or were recently acidic (Stephensen and Mackie 1986). These investigators also determine the acute toxicity of H^+ for adult Hyalella; the four and ten day LC_{50} values were 4.4 and 4.5 respectively. France and Stokes (1986) found that the LC_{50} increased with decreasing size/age and was dependent on the molt cycle state. Symptoms of toxicity appeared over eight days at pH 5.5 and below. These laboratory results suggest that Hyalella may be susceptible to episodes of low pH, although autumnal events, when the population is composed primarily of young animals, are most likely to have an impact. Field observations, however, suggest that reproductive impairment may occur at a much higher pH (e.g. 5.6). For example, in Plastic Lake (mean alkalinity 7.8 μeq/L, mean pH 5.68) only a few animals were found in 1984 (Stephenson and Mackie 1986). In Heney Lake (Mean alkalinity = 8.2, pH 5.67) a two week reproductive delay was observed in 1983. Both mechanisms may affect the populations.

Some species respond positively to lake acidification, and in some cases the effect is detrimental from a human use standpoint. Blooms of an odour forming alga, Chrysochromulina breviturrita is a well studied example. In recent years it has been collected from over 80

acidic and softwater lakes in Ontario, but never in alkaline lakes (Nicholls et al. 1982). The pH preference of this alga has been studied in pure culture, and the pH at which blooms have occurred in nature (5.5-6.2) correspond to the middle portion of its allowable pH range. Good growth in laboratory cultures occurs over a wider pH range (4.0 to 6.9) than in lakes, probably because other factors have been optimized. A selenium requirement has been demonstrated for this alga, and episodes of atmospheric deposition of selenium originating from coal-fired power plants may explain the unpredictable nature of Chrysochromulina blooms (Wehr and Brown 1985).

Other nuisance algae associated with acidification are the slimy, filamentous forms that bloom in the littoral zone of slightly acidic lakes. Three lines of evidence indicate that these blooms are caused by acidification. Firstly, in a series of limnocorrals in Lake 223, lowering the pH from 6.5 to 6.0 caused an increase of Mougeotia from 19 to 61% of the periphyton biomass. Other filamentous forms also increased, especially at lower pH's such that together they comprised 70-80% of the total biomass between pH 6.0 and 4.0 (Muller 1980). Secondly, acidification of Lake 223 led to the development of thick mats of Mougeotia in the littoral zone (Schindler et al. 1985). Thirdly, neutralization of Bowland Lake eliminated extensive nearshore accumulation of Mougeotia and Zygogonium (Jackson in press).

The biological significance of the blooms of filamentous algae is unknown, but the recreational value (swimming, aesthetics) of affected lakes is impaired.

Astrionella ralfsii is a large diatom that bloomed in Lake 223 at pH 5.6 and below (Schindler et al. 1985). Unlike the algae discussed above, it does not affect the water quality from a human use standpoint, nor has any direct biological consequence been demonstrated. Nevertheless, a few adverse effects can be inferred. The frustules of this species are resistant to dissolution and sink rapidly to the sediments, carrying with them silicate and possibly other nutrients. A decline in soluble silicate was associated with the blooms of A. ralfsii in Lake 223 (Schindler et al. 1985). The decrease in silicate may be sufficient to competitively inhibit the other major species of diatoms, in particular Cyclotella spp. Because the latter are highly edible by zooplankton but A. ralfsii is not, the blooms of this species may decrease the edible fraction of phytoplankton in lakes.

There are indications that several other species may be adversely affected at relatively high pH's (5.5-6.0), but because the data are incomplete they will be considered only superficially.

Amongst the snails, Servos (1983) found Campeloma decisum, Gyraulus parvus, and Helisoma anceps in 65% of 16 lakes above pH 6.3, but their respective frequencies of occurrence were 50, 25 and 19% in 16 lakes between pH 5.5 and 6.2. Many laboratory bioassays have been performed with aquatic insect larvae and nymphs (summarized by Singer 1984) and the long-term survival of several species is affected between pH 5.5 and 6.0. Unfortunately, survey data for these species are unavailable, so the significance of the laboratory data is not clear. Hall and Ide (1986) compared historical and recent data on stream insects and found that a number of species of mayflies and stoneflies have disappeared from two sites subject to spring pH depressions. Because an equal number of different species replaced the missing species, the ecological implications of these data are at present unclear. In Norway, the amphipod Gammaris lacustris disappears at 5.8 to 6.0, and even though this species occurs in North America no information on its frequency of occurrence or abundance in North American softwater lakes is available. Two diatoms, Synedra acus and Achnanthes minutissima comprise a major portion of the spring and early summer periphyton biomass (typically 25% each). These species dropped to very low levels at pH 6.0 and below in Muller's (1980) limnocorral experiments. The significance of these results is unclear due to the lack of survey data and information on their ecological role.

The above data on sensitive species has been summarized in Tables 5 and 6 along with the information on several tolerant forms.

Table 6. The pH Levels at which Various Aquatic Organisms Respond Positively to Acidification.

Critical pH	Species	Comments	Reference
6.0-4.0	Mougeotia and other filamentous algae	Field experiments	Muller 1980 Schindler et al.
5.5-6.2	Chrysochromulina brevitturita	Laboratory experiments and field observations	Wehr et al. 1985 Nicholls et al. 1982
5.5-5.0	Asterionella ralfsii	Field experiment	Schindler et al. 1985

Table 5. **The pH Levels at which Various Aquatic Organisms Respond Negatively to Acidification.**

Critical pH	Species	Sensitive Life Stage or Event	Comments	Reference
6.0	*Gammarus lacustris*	No data	Norwegian field survey	Økland & Økland 1985
6.0	*Achnanthes minutissima*	No data	Field experiment	Møller 1980
6.0	*Synedra acus*	No data	Field experiment	Møller 1980
5.6-5.9	*Mysis relicta*	No data	Field experiment	Nero and Schindler 1983
5.6-5.9	*Orconectes propinquus*	Stage I-III juveniles	Lab and field experiments	Berrill *et al.* 1985
5.6-5.9	*O. rusticus*	Stage I-II	Lab and field experiments	Berrill *et al.* 1985
5.6	*O. virilis*	Juveniles	Field expriment	Davies 1986
5.0-5.5	*Amnicola limnosa*	Embryonic development	Laboratory experiments	Servos *et al.* 1985
5.6 (?)	*Hyalella azteca*	Probably juveniles	Field observations	Stephenson and Mackie 1985
5.0-5.6	*Daphnia galeata mendotae*	No data	Field experiment	Schindler *et al.* 1985
4.1-5.0	*Daphnia pulex*	Reproduction	Laboratory experiments	Walton *et al.* 1982
4.5-4.8	*Cryptomonas ovata*	No data	Laboratory experiments	Bowen and Ward 1978 Moss 1973

3.3.3.1 Alkalinity requirement for protection of non-fish biota

Although both laboratory and field data could be utilized in determining an acceptable pH, two complicating factors indicate that only field data should be used. Firstly, laboratory results tend to underestimate the toxicity of substances in the field, probably because other stresses are eliminated in the laboratory and because field chemical data are usually reported as means. Extreme values may be more important. Secondly, the relationship between alkalinity and pH in a lake often deviates significantly from theoretical expectations. Thus, even though organisms respond to hydrogen ion, conversion of laboratory pH to lake alkalinity (upon which acidification models are based) is fraught with difficulties.

The most compelling results are certain extinction events (or reproductive failures) that occurred in Lake 223, Plastic Lake, and Heney Lake. The average pH and alkalinity of these lakes on or around these events are presented in Table 7. Most of the *Mysis* population was lost between August of 1978 and August of 1979, and 1979 was the first of several years of reproductive failure of *Orconectes virilis*. These results suggest that an epilimnetic alkalinity of 5.5 μeq/L and pH of 5.64 will not protect these two species. Because acidification of Lake 223 was so rapid the survival of *Mysis* at 12 μeq/L and pH 5.93 is ambiguous. *Amnicola* and *Hyalella* were lost at epilimnetic alkalinities of 3.7 and 6.1, respectively, and mean pH of 5.8. Clearly, epilimnetic alkalinities of 4 to 6 μeq/L are inadequate to protect these species. Minimal "safe" values can be inferred from the year prior to reproductive failure. This appears to be 12 μeq/L for *Orconectes*, 12 to 17 μeq/L for *Mysis*, and 10 μeq/L for *Hyalella*. The appropriate data for Heney Lake are unavailable. However, reduced production and growth rate of this species were observed in two lakes with alkalinities of 8.2 and 13.1 μeq/L (Rooke and Mackie 1984b). The range of all these "safe" values is 8 to 17 μeq/L. Thus an epilmnetic alkalinity of 15 μeq/L would appear to be the borderline below which adverse effects occur. Fluctuations of deposition and hydrologic regimes may cause year to year variations of up to 10 μeq/L. Thus, to provide protection from "worst case" annual fluctuations, a minimum alkalinity of 25 μeq/L is required. A small margin of safety is desirable, e.g. 5 μeq/L. Hence, the minimum alkalinity recommended for protection of non-fish biota is 30 μeq/L.

3.3.4 Evidence of Effects of Acidification on Aquatic Dependent Wildlife Communities

3.3.4.1 Aquatic Birds

Due to their dependency on the aquatic environment for nest sites, brood protection, and food, aquatic birds may be indirectly affected by the impact of acid precipitation on aquatic biota. The severity of

Table 7. Average pH and Alkalinities Associated with Documented Reproductive Failures. Note: Whole lake values were time and volume weighted. Sources: Staff of the Department of Fisheries and Oceans, and Ontario Ministry of the Environment.

Lake	Year	Epilimnetic		Whole Lake		Event
		pH	Alkalinity (μeq/L)	pH	Alkalinity (μeq/L)	
223	1977	6.13	16.6	6.21	12.9	
	1978	5.93	12.0	5.87	19.3	Possible stress on *Mysis*
	1979	5.64	5.5	5.63	13.2	Demise of *Mysis*, Partial reproductive failure of *Orconectes*
	1980	5.59	0.8	5.50	6.9	Complete reproductive failure of *Orconectes*
Plastic	1983	5.79	6.1	5.68	8.2	Reproductive failure of *Hyalella*
Heney	1980	5.96	3.3	5.61	8.1	Reproductive failure of *Amnicola*

such effects will be determined by the sensitivity of the habitats in which nesting territories are established and where young are raised. Early brood-rearing is the critical life-stage when many species of aquatic birds are obligate fish or invertebrate feeders and may be affected by changes in prey availability in the vicinity of the nest.

Recent findings indicate that breeding success and growth of young may be lower in highly acid-sensitive streams and headwaters where essential prey organisms are limited by nutrient availability as well as low pH. A report of temporal change in aquatic bird numbers has been published for Wales where a decline in the abundance and breeding success of dippers (Cinclus cinclus) over the past 20 years has been found coincident with increased stream acidity and reduced densities of aquatic insect prey organims.

In central and northern Ontario, spatial comparisons of water fowl breeding success have shown that productivity is significantly lower in the headwater lakes receiving deposition greater than 30 kg.SO_4/ha/yr based on a large sample of lakes in two areas (McNicol et al. 1985). While physically similar, the study areas differ dramatically in lake acidity. More than 66% of the Wanapitei lakes were acidic (pH $\angle$ 5.5), compared to less than 10% in the Ranger Lake study area. All study lakes exhibited low alkalinities, although water sulphate concentrations were significantly higher at Wanapitei (Ibid).

The waterfowl breeding success rates found in this study indicate that the abundance of fish stocks and the structure of fish communities observed at moderate pH levels (pH 5-6 range) have a significant impact on the breeding capabilities of aquatic birds (McNicol et al. 1985). Although the selection of lakes varied for piscivorous and insectivorous waterfowl, the utilization of acid-fishless lakes was somewhat less than expected for all waterfowl species in central Ontario (Ibid). A comparison of the percentage of lakes occupied by broods, in relation to the percentage occupied by breeding pairs, suggested a substantial difference in waterfowl production between the two study areas.

Despite the pronounced differences in chemical and trophic conditions between the two study areas, no significant differences were observed in the distribution of nesting waterfowl among the various lakes studied during spring breeding pair surveys. Yet a much smaller percentage of lakes supported broods of all species at Wanapitei than at Ranger Lake. The ratio of broods to the breeding pair numbers on the study lakes in the Ranger Lake area was approximately 1:2 for loons and mergansers, and close to a 1:1 ratio for insectivorous waterfowl (e.g. ring-necked ducks, black ducks and mallards). In the acid-stressed Wanapitei area, however, this ratio was generally lower for all birds studied but particularly for the fish-eating waterfowl which were found to have a very low success rate of 1:17 (McNicol et al. 1985).

This discrepancy between the number of nesting pairs and young produced between areas suggests reproductive failure following nest initiation (McNicol et al. 1985). It is speculated that the failure is due to reduced food availability in the acidic fishless lakes as compared to the naturally fishless lakes found in the control area (Bendell and McNicol in press).

No differences in diet were apparent for ring-necked ducks collected from lakes with or without fish, while common goldeneyes from fishless lakes had a greater proportion of odonates, trichopterans, hemipterans and coleopterans than those from the lakes with fish (Bendell and McNicol in press).

The diets of some fish and ducks overlap extensively in simple headwater lake systems, where competition occurs for the same food resource (Eriksson, 1979; Eadie and Yeast 1982; Hunter et al. 1985). It has been shown that fish and ducks will compete for similar prey organisms in acid-stressed lakes, because fish are usually not eliminated by moderate increases in acidity (Desgranges in press).

The critical life-stage for food availability is the early brood development period covering the first six to nine days following hatch. During this period, many species of aquatic birds are obligate invertebrate feeders and compete with fish for availabe aquatic insect prey (i.e. in the water column).

Growth rates of black duck and common goldeneye ducklings were monitored in the field to assess feeding requirements and behavior in relation to food availability under acid-stressed conditions and in the presence of competition with fish (Desgranges in press). On a neutral lake with fish, the growth rate of black ducks was 7.4 g/day compared to the growth rate on an acidic lake without fish of 9.4 g/day, and the duckling growth on an acid lake with fish of 3.8 g/day. Similarly, imprinted goldeneye ducklings maintained good growth rates on both the neutral lake with fish and the acid lake without fish, however, growth was greatly reduced on the acid lake with fish. Comparisons of growth rates recorded in an earlier study in Maine by Hunter et al. (1985) followed the same pattern of higher black duck growth rates on the acid lake without fish (4.8 - 5.3 g/day) as compared with the neutral lake with fish ($<$ 3.3 g/day). However, the effects on growth of the combined conditions of acidity and competition with fish were not tested in this study.

The results of an experimental field study where shallow emergent wetlands were acidified and where imprinted ducklings were reared (Harrimus 1984) confirm the finding that chemical alterations causing disruption to invertebrate community structure (i.e. below pH 5.5) greatly reduce duckling growth and survival. Ducklings placed on the acidified ponds spent more time searching for food and gained significantly less weight than those reared on the unacidified ponds (Ibid).

Temporal trends in numbers of breeding birds in response to an increase in stream acidity has been reported in upland Wales (Ormerod et al 1985). The breeding distrbution of dippers (Cinclus cinclus) was studied in relation to the abundance of important prey organisms and changes in stream acidity over recent decades (Ibid). Densities and distribution of these birds were strongly correlated with the abundance of larval trichopterans and nymphal ephemropterans which are important prey for dippers while feeding nestlings. These aquatic insects were scarce in acidic streams as compared to earlier surveys of their abundance. Over the past 20 years, the stream acidity has increased while aquatic insect and dipper populations associated with these streams have declined (Ibid).

3.3.4.2 Amphibians

There is a large body of evidence from field and laboratory experiments that shows that elevated hydrogen ion concentrations are highly toxic to amphibians (Gosner and Black 1957; Pough 1976; Saber and Dunson 1978). There is good evidence to suggest that poor reproductive success is a factor resulting in smaller populations in very acidic habitats. Increased embryonic mortality at low pH (pH < 5.0) has been measured in both natural populations as well as in situ bioassays (Saber and Dunson 1978; Clark and Hall 1985; Clark 1986; Freda and Dunson 1986; Dale et al. 1986). Freda and Dunson reported extremely high mortality (62 to 100%) of Ambystoma jeffersonianum embryos in in situ bioassays in five of six acidic ponds uninhabited by this species.

The pH levels at which increased embyronic mortality occurs varies greatly among species. Many amphibians are not affected unless the pH falls below 4.6 (reviewed by Freda in press). However, the salamanders are more senstive; for two species native to Canada that have been tested, embryonic mortality can occur at pH < 5.0 (Ibid).

At extreme pH levels (< 4.5) embryonic development is immediately arrested (Pough and Wilson 1977), Pierce et al. 1984). At less toxic levels, hatching is inhibited and developmental abnormalities of hatched larvae are common (Dunson and Connell 1982; Freda and Dunson 1985a; Clark and LaZerte 1985).

Aluminum concentrations which are often elevated in acidic natural waters are also toxic and can further reduce embryonic survival (Clark and LaZerte 1985; Clark and Hall 1985; Glooschenko et al. 1985). As little as 10 g/L at pH < 4.6 increases mortality of Bufo americanus embryos under laboratory conditions (Clark and LaZerte 1985).

Larvae are more acid tolerant than embryos, but are more vulnerable to the indirect effects of acidification because they are feeding very actively during this life stage; food resources and competitive relationships are very important. Community structure and species compositon of the food resources of larvae, mainly benthic invertebrates and plankton, can be altered at pH < 5.6 (Mierle et al. in press). Larvae have reduced growth rates at low pH and body size influences competition and predator-prey relationships (Wilbur 1984; Freda and Dunson 1985b, 1986; Ling et al. 1986).

Indirect evidence from distributional data show that amphibian populations are restricted in acidic habitats. Studies in Canada, the United States, and Europe have reported reduced amphibian populations in ponds with low pH (pH < 6.0) (reviewed by Freda in press). In central Ontario in ponds with pHs from 4.55 to 6.37, Ambystoma maculatum, Hyla crucifier, Rana catesbeiana, Rana clamitans and Rana sylvatica were less common in the more acidic ponds (Clark 1986 in press).

There are 17 amphibian species in Canada whose ranges overlap with areas being affected by acid precipitation. Most of these species breed in beaver ponds, temporary meltwater ponds and small lakes. Many of these aquatic habitats are more susceptible to acidification and acid shock events than are larger lakes because they have high flushing rates and low buffering capacity.

A number of surveys of amphibian breeding habitats have been conducted in Canada outside of areas infuenced by local sources of contamination. The results indicate that many amphibian habitats are very acidic, with a large variability in the contributions of organic acidity (Pough 1976, Dale et al. 1985, Gascon and Planas 1986, Clark in press). In central Ontario, 13% of 60 ponds studied were at pH levels that have been reported to reduce hatching success of some amphibian species (pH < 5.0) and another 33% of the ponds sampled were at levels that are highly susceptible to short term pH depressions (pH 5.0 to 5.5) and where larval food resources could be impacted (Clark 1986, Clark in press).

By way of comparison, 13% of the 31 ponds studied in Nova Scotia were found to have pH values less than 5.0 and 7% were between pH 5.0-5.5 (Dale et al. 1985), while 24% of the ponds studied in the Sudbury area had pH values < 5.0 and 6% of the ponds had pH values between 5.0-5.5 (Glooschenko et al. 1985), although ponds studied in these two areas were located by listening for calling frogs, a potential bias in the sampling efficiency of amphibian habitats. Hyla crucifer occurred in fewer of the low pH ponds (within 60 km of Sudbury) than in the higher pH ponds.

Amphibians form an important link in the flow of energy between aquatic and terrestrial ecosystems. In aquatic habitats, adults and larvae amphibians are major consumers of algae and invertebrates and are important food items for invertebrates, fish, birds and mammals (Dickman 1968; Debenedictis 1974; Seale 1980; Racey and Euler 1983). In terrestrial ecosystems, adult amphibians are a high quality source of protein, can be important contributors to biomass (Burton and Likens 1975), and channel energy from the aquatic to terrestrial environment (Wassersug 1975).

3.4 What evidence is there of a relationship between the chemical changes and the biological status of the aquatic ecosystem?

3.4.1 Established Linkages between Aquatic Chemstry and Fish Communities

The best evidence of linkage between pH decline in lakes and fish population decline is from two whole-lake manipulation experiments at the Experimental Lakes Area. The pH of lake 223 was first lowered from 6.8 (1976) to 5.1 (1981), and then maintained at 5.1 (1981-1983) (Schindler et al. 1985). The slimy scuplin and fathead minnow became extinct. Abundances of lake trout, pearl dace, and white sucker increased during the early years of acidification and decreased during the later years of acidification. The abundance declines of all five species were due primarily to recruitment failures. Increased mortality of adult lake trout, due to food web disruption, could account for 1/3 of the population decline, but the primary loss mechanism was recruitment failure (Mills et al. 1986). Lake 304 was acidified for three years and the pH declined from 6.4 to 5.2. Fathead minnow and red belly dace populations disappeared (Schindler and Turner 1981; Mills and Schindler in press).

Other evidence of linkage between pH change and fish population modification come from stream manipulation experiments, and from lake liming projects. In a stream acidification experiment in the Hubbard Brook Experimental Forest (Hall et al. 1980) most brook trout moved away from the acidified section. No mortality was observed. Whole lake addition of calcite to an acidified former lake trout lake, Bowland Lake, improved water quality such that stocked 0^+ and 1^+ lake trout have survived and exhibited good growth. Transferred adult lake trout spawned both autumns following neutralization (Booth et al. in press). Improved survival of lake trout eggs and fry held in situ suggest that if the current water quality is maintained the stocked lake trout population may reproduce successfully (Booth et al. in press).

3.4.1.1 The relationships between fish body chemistry and lake conditions

In aquatic systems undergoing acidification, coincident chemical changes can occur to the point where catastrophic changes result in fish populations. These chemical changes may also lead to either demineralization of bony tissue or accumulations of metals whose concentrations are exacerbated by declining pH. In all cases, the inferential linkage between chemical conditions and the elements exists; however, fish growth, individual species lifestyles and lake watershed features appear at varying times to influence the direct relation between pH and body concentrations. Table 8 is a summary of some significant studies that have been reported.

Table 8. Reports of Fish Body Chemistry in Relation to Habitat Chemistry.

Reference	Result
Fraser and Harvey 1982	Bone decalcified and Mn increased in bone, six lakes studied central Ontario.
Wren and MacCrimmion 1983	Hg body burdens higher at lower pH but a function of lake size, depth and Ca^{2+} for pumpkinseed.
Kelso and Gunn 1984	Body burden can be described by multiple regression for brook trout and white sucker but regression components (pH, fish size, lake morphometry) differed by species. Growth was an important factor.
Moreau *et al*. 1983	Mu, Zn, Sr higher in opercula and scales of fish from acidified lakes.
Richard *et al*. 1985	Al on gills and Zn and Mn in bone directly correlated to acidity.
Kelso *et al*. 1986	Hg in species from eastern Canada correlated to lake morphometry characteristics and pH in some regions.
Suns *et al*. in press	Epilemnetic pH inversely correlated with Hg burdens of young of the year yellow perch. Also Hg related to fish condition.

3.4.2 What Evidence is there for a Relationship between Food Chain Declines and Fisheries Response?

While the impact of food web degradation for accelerating the loss of fish populations has received little attention from researchers, the lake 223 experiment shows that food web disruption did lead to decreased growth and condition of lake trout, and was accompanied by an increase in adult mortality (Mills 1984). This indicates that this change may be important for some aquatic systems.

3.4.3 What is the Evidence that Episodic Acid Shock Events are Critical to the Biological Community?

The storage of acidic ionic species in the accumulated snow pack and the concentration of these species in the early phases of snowmelt are well documented in the literature. This phenomena is summarized in a workshop report by Marmorek et al. (1984) and has been measured in many areas. See Cadle et al. (in press), Scheider et al. (in press), English et al. (in press), Semkin and Jeffries (in press), or Jones et al. (in press). It has been demonstrated conclusively that tributary streams and waters of the surface and littoral zones of lakes suffer severe depletion of alkalinity, increases in acidity, and increases in free aluminum for periods of several days during the snowmelt. The ability to simulate the occurrence and intensity of this "acid shock" is advanced and reasonable validations of models has been achieved. Jones et al. (in press), Lam et al. (in press) Rustad et al. (in press), and Goodison (in press).

However, the evidence of the importance of this episodic, short period decline in water quality to biota remains under active investigation. Harvey and Welpdale (in press) have shown that rainbow trout lost plasma, Na and Cl from body mass and died in 28 hours when exposed in cages in a natural spring melt water. Gunn and Keller (in press) have measured the occurrence of such lethal conditions in spawning sites which would indicate a severe restriction of available spawning habitat. Gunn and Noakes (in press) have shown in experimental tests that sac fry avoid the low pH, high aluminum environment if given the opportunity.

Actual documentation of mortality of wild stock during the period of severe acid shock events are rare but have been observed in Plastic Lake, Ontario (Harvey 1984). However, the extensive research findings, cited above, clearly indicate that conditions occur in headwater streams and in the shallow littoral and surface zones of lakes that would be unfavourable and perhaps lethal to survival of important biological species. The existing adult stock may avoid these conditions but are thereby denied important segments of habitat. For other life stages such avoidance may not be possible. These conditions may result in differential mortality of younger classes (Frenette and Dobson 1984). It would appear that evidence now

exists that major segments of headwater streams are subjected to episodic periods of increased acidity and elevated aluminum concentrations that reach lethal or near lethal levels. Such conditions also likely occur in important segments of many lakes that have more acceptable water quality during the balance of the year. The importance of episodic acid pulses to amphibian populations is uncertain. We do know that the most sensitive developmental period of many amphibians, the embryonic life stage (Gosner and Black 1957; Pough 1976; Freda in press), can coincide with the time of snowmelt. It has been demonstrated that mortality of embryonic amphibians is caused by short-term exposures to low pH (Tome and Pough 1982) and low pH in combination with elevated concentration of aluminum (Clark and Hall 1985; Dale and Freedman 1985).

The total implication of the short-term acidic events on different segments of the biological community is not fully understood nor are the importance of recurrence or timing in relation to species life cycles sufficiently established. The potential for serious population influences clearly exists.

3.5 Can the rate of response and rate of return to equilibrium under a new level of deposition stress be defined quantitatively?

Evaluation of the rate of ecosystem response to acid deposition and whether this response is reversible remains among the most difficult aspects of current LRTAP research. When intensive research into acid deposition effects was initiated, insufficient reliable information was available on historical conditions and the degree of natural variability in water quality on which to estimate the rate of change in acidity of surface water systems; to a certain degree this is still the case.

Multi-year LRTAP studies have been conducted in some locations in eastern Canada and the time series data collected within them is now beginning to yield clues to the reaction rate and reversibility questions. In Nova Scotia, where a decrease in the already moderate level of deposition over a 5-10 year period has resulted in decreased sulphate concentrations in surface waters with coincidently increased pH, and therefore alkalinity (Thompson in press). Substantial decreases in deposition near Sudbury over a 5-10 year period due to a combination of technological and economic factors has resulted in a similar increase in lake pH's (Keller et al. in press; Hutchinson and Havas in press; Dillon and Girard in press). Similar indications of the capacity for reversing lake acidification have been reported in Sweden (Forsberg et al. 1985). Finally, the RAIN project in Norway (Wright et al. in press) showed that increased deposition to a previously unaffected, sensitive terrestrial catchment resulted in

rapid and significant changes in outflow chemistry (increased acidity, sulphate, ntirate and labile aluminum). Reduction in deposition to a previously acidified terrestrial catchment has resulted in a small but immediate reduction in the outflow acidity and sulphate levels and a larger reduction in nitrate. All of these studies suggest that for the Canadian Shield terrain typical of most of Eastern Canada, a fairly rapid reversal in acidification (i.e. years rather than decades) can be expected in response to a decrease in acidic deposition.

Because of the lack of reliable long-term monitoring records, the estimation of long-term responses depends on using the available data for development and validation of simulation models. This requires measurements of cation weathering rates that are most difficult because of the spatial variability and the slow interactions (Skeffington and Brown in press; and Riha et al. in press). The treatment of the aquatic responses to weathering rate has often been oversimplified in process-oriented models. In general, these formulations are hypotheses at best. The work by Wright et al. (in press) has indicated that it is difficult to discern the cause-effect of weathering and that the precipitation sulphate plays the controlling role in watershed acidification. In particular, Cosby et al. (1985) has to choose the appropriate weathering rates from a fairly narrow range in order to obtain a good simulation of the present streamwater chemistry at White Oak Run in a 140-year simuation. That is, weathering rates could be bracketed reasonably closely by using current observations (Reuss et al. (1985). In the case of the ILWAS model (Gherini et al. 1985), the weathering rate is assumed proportional to $[H^+]^a$, where the power a lies between 0.3 and 0.7. It appears that the dependence of weathering rate on pH probably exists, but whether this functional dependency is correct is not known. A similar power law is also proposed by Schnoor and Stumm (1985) in the Trickle-Down model. Others have proposed that weathering rates should be formulated as fractional order with respect to soil solution acidity instead of the power law (Reuss et al. 1985). Still others (e.g. Arp 1983) relate the weathering rate directly to the acid deposition and assume it to be independent of base content in the soil.

Intensive geological surveys have helped in defining the rock minerals and their weathering rates (e.g. April et al. 1985; Likens et al. 1977; Kramer et al. 1981). A comprehensive analysis of such observed data using the mass balance is best illustrated by the work of Paces (1985). Craig et al. (in press) also related the weathering rate to hydrological parameters such as soil contact time. It is, however, difficult to define the weathering rate to a higher degree of accuracy than these attempts. More experimental work is therefore required before a high level of confidence can be placed on model simulations over extended periods.

The one critical concern associated with the rates of response is the rate at which recovery will be realized when the deposition is reduced. It is clear by now that the recovery function differs if the watershed hydrological and hydrogeochemical characteristics differ. A good demonstration has been provided by Galloway et al. (1983) for several scenarios in which the acid deposition is changed. Parts of these scenarios are shown to be realistic by Wright et al. using the diatom data for several lakes (in press). The work by Dillon and Girard (in press), and Johnson et al. 1985 on Sudbury area lakes also confirmed the necessity of using an F-factor in the CDR model to provide the correct recovery. In the U.S. National Acid Precipitation Assessment Program (Bennett in press); Malanchuk et al. in press), simple empirical correlations are used to define direct and delayed responses in aquatic regimes. For example, for lakes with negative alkalinity, the impact of acid deposition is direct. At the other extreme, the watershed can be under a capacity protected condition if the alkalinity is more than 400 μeq/L. In between these two extremes, more sophisticated models are used to define the delayed response time, using the concepts of soil contact time, percentage base saturation and sulphate adsorption isotherms. Even more sophisticated models (e.g. ILWAS, MAGIC, and Trickle-Down models) may then be used to define the more difficult cases. A great deal of extrapolation is required to define the regional scale of responses (Malanchuk et al., in press).

3.6 What is the Potential and Associated Possible Consequences of Mitigative Measures?

The chemical consequences of lake and stream liming have been well established through the extensive programs conducted in Sweden (Sverdrup 1985). Three independent liming studies have been undertaken in Canada; two in Ontario were directed at restoring or protecting trout populations in acidified lakes, and one in Nova Scotia was aimed at restoring acidified Atlantic salmon habitat in rivers.

3.6.1 The Sudbury Lakes Environmental Study

Three highly acidic, metal-contaminated lakes near Sudbury, Ontario, were limed by the Ontario government beginning in 1973 with a combination of calcium hydroxide and calcite in an attempt to restore fish (Yan and Dillon 1984).

Copper and nickel levels in lake waters were extremely high prior to liming because of metal deposition from nearby smelting operations. Although liming resulted in major increases in pH (to pH > 7) and reductions in metal levels, the waters remained highly toxic due to

residual metals (Yan and Dillon 1984). As input of acid and metals continued, the lakes began to re-acidify and metal levels increased to their original levels.

Two of the lakes were fertilized with several annual additions of phosphate after liming (Yan and Lafrance 1984). Although fertilization of limed lakes showed some promise as a tool to delay reacidification in the Sudbury study, further research and modelling efforts are required before deciding whether fertilization is a useful management tool for neutralizing acidified lake waters.

3.6.2 Experimental Lake Neutralization in Ontario

In Ontario, the Ministries of Natural Resources and the Environment have undertaken (in 1981) an Experimental Neutralization Program to assess the feasibility of using lake liming to rehabilitate acidified lakes and to protect the biological communities in acid-stressed lakes. The program is still underway but some preliminary findings have been published (Booth et al. in press).

In 1983 neutralization of Bowland Lake, an acidic lake near Sudbury, Ontario, that formerly supported a viable lake trout population, raised the whole lake pH from 5.0 to 6.8 and resulted in reduced concentrations of total aluminum (from 130 to 65 $\mu g/L^{-1}$).

Young lake trout (0^+, 1^+) stocked postliming, survived and exhibited good growth. Transferred adult lake trout spawned in both years after liming. The resident yellow perch population showed both increased growth (older cohorts) and decreased growth (younger cohorts) and a four-fold increase innumbers. These responses of the yellow perch populations are not yet attributed to the liming.

The long-term response of Bowland Lake fish populations is yet to be determined, but chemical models suggest that, if it is not re-limed, Bowland Lake will reacidify to pre-neutralization water quality by 1990. The lake trout population may be in jeopardy much earlier.

A low alkalinity lake, Trout Lake, near Parry Sound, Ontario, was limed in 1984. No immediate response of the fish community was observed; however, the community did not appear stressed prior to lake neutralization. Booth et al. (in press) did observe 100% mortality of lake trout fingerlings held in cages at one nearshore site post-neutralization during spring melt, suggesting whole lake liming was not entirely effective in mitigating episodic pulses of snowmelt at all littoral sites.

As whole-lake neutralization may not protect critical spawning habitat, Ontario is experimenting with shoal liming (addition of

limestone gravel to spawning shoals) as an alternative or addition to whole lake liming.

Ontario's current approach to neutralization is an experimental one. The province remains committed to abatement of emissions at source as the only solution to acidic deposition. Until the long-term response of fish communities to neutralization has been assessed, lake liming will not be used on a widespread basis.

3.6.3 Salmon River-Liming in Nova Scotia

Long range transport of sulphuric acid has caused the extinction of Atlantic salmon (*Salmo salar*) stocks in 13 Nova Scotian rivers and severe declines in an additional 18 rivers (Watt *et al.* 1983; Watt 1985). Fearing further losses, the Department of Fisheries and Oceans has undertaken experiments (Watt *et al.* 1984; White *et al.* 1984) to test the feasibility of mitigating the acidification of Atlantic salmon rivers in Nova Scotia by addition of limestone or other substances to lakes or streams. Two liming methods have been tested extensively, instream limestone gravel (at six river treatment sites) and headwater lake liming (in five lakes), and estimates have been made of the relative costs and effectiveness.

Three years of observations at six sites where limestone gravel had been placed in the streambed produced disappointing results (Watt et al. 1984) and led to the conclusion that this approach is impractical (Watt 1985). The results can be summarized in the form of the following equation:

$$\Delta pH = 0.237 (\ln DOSE) + 0.008 (^{\circ}C) - 0.809$$

where ΔpH represents the rise in pH that can be expected for a given limestone gravel DOSE (in metric tonnes of gravel per cubic meter per second of stream discharge) and water temperature. Instream pH was not a variable; all six sites had mean annual pH's near 5.0. Low water temperatures during winter and early spring reduced the rate of limestone dissolution, and at high flows the amount of limestone gravel that would theoretically be required to give a pH increase of as little as 0.3 units was so large as to be prohibitive.

The second approach has been the liming of headwater lakes to create a reservoir of treated water, which is metered out by natural discharge from the lakes to protect downstream salmon habitat. The limestone was usually added to the lakes as a slurry sprayed evenly over the entire surface. Watt *et al.* (1984), in a four lake study, have shown that, even with lakes of mean residence time as short as five months, satisfactory lake pH levels can be obtained for up to one year if the limestone dosage exceeds three times the whole-lake acidity. The

overdosing provides an excess of lime which settles to the bottom, thus effectively sealing off acid demand from the sediments, and dissolving slowly to provide additional alkalinity at each fall and spring turnover. The treatment must be repeated annually in lakes with mean residence times of less than one year. Calcitic limestone was found to give better dissolution efficiencies than the dolomitic variety.

A major problem with the headwater lake liming approach, as noted by White et al. (1984), is that during episodes of heavy autumn and winter rains the lakes can become covered by a thin surface layer of highly acidic rainwater which then flows out of the lake to deliver a downstream acid shock. This phenomenon is most common under ice cover when an inverse thermal stratification prevails (Watt et al. 1984). Watt (in press) reports that this problem has been overcome by spreading a layer of dry limestone powder evenly over the ice surface. This form of liming is effective in preventing the buildup of an acid surface layer because the first water entering the lake from a winter rainstorm is drainage from the ice cover.

Positive responses from Atlantic salmon were obtained with both the instream-gravel and headwater-lake liming methods (Watt et al. 1984). Significantly higher densities of juvenile salmon were found in the immediate vicinity of instream limestone gravel deposits, but this beneficial effect did not persist far downstream. In the lake liming studies, salmon parr introduced into the previously barren and toxic outlet stream of Sandy Lake, were still surviving one year after liming. In addition, wild native salmon adults migrated (apparently attracted by the higher pH's) into the previously unused outlet stream and spawned successfully. The resulting wild salmon fry were showing good survival up to one year after liming.

The total estimated cost (Watt in press) for a 20-year project of de-acidifying the Atlantic salmon habitat of the Southern Upland of Nova Scotia would be 95 million dollars (1984 $ Canadian). The aim of such a full scale liming effort would be to bring the Atlantic salmon production level of Nova Scotia's acidified Southern Upland back up to the pre-acidification production level, estimated at 45,200 adult salmon (total stock) per year. The actual annual Canadian catch (sport and commercial) from this production area would be about 24,000 salmon, which represents an enhancement potential of 12,000 salmon above the present average catch, at an average annual cost of 4.75 million dollars over 20 years. These costs amount to approximately $400 per landed salmon. The value per landed salmon to the eastern Canadian economy is, on average, less than $100 per fish (McPhee et al. 1981), hence the full-scale liming operation described above cannot be justified in economic terms.

A large area of Nova Scotia has already been rendered barren of Atlantic salmon, and more is expected to be lost over the next 20 years. After the acid rain problem is brought under control, it will be necessary to mount a salmon restoration program for the barren habitats. The future job of restoration would be easier and entail a higher probability of success, if a number of nuclei of native wild local stocks were available to provide a genetically diverse selection of potential donor stocks. To achieve such a gene bank, Watt in press recommends that a number (approximately six) of the 18 river stocks currently threatened with extinction should be preserved by using the headwater lake liming technique to create de-acidified refuges in tributaries.

In summary, the lake and river liming experiences in Ontario and Nova Scotia highlight the problems with full-scale liming operations as a means of rehabilitating and protecting aquatic ecosystems:

1) in highly metal-contaminated systems, liming may not reduce metal concentrations to non-toxic levels;
2) without concommitant reductions in deposition, liming is a continuous process;
3) liming may not protect the habitat for critical developmental periods of all fish species;
4) the long-term response of resident fish populations to liming has not yet been assessed;
5) as liming is not an exact reversal of the acidification process and has been studied in a limited number of systems, we cannot predict the response of the resident community;
6) on strictly economic terms, full scale liming of Nova Scotia Atlantic salmon rivers cannot be justified.

3.7 Aquatic Resources-at-Risk in Reltion to Acidic Sulphate Deposition

The aquatic resource-at-risk in relation to the effects of acidic deposition could be defined in terms of the actual total biosystem utilization of the water resource. This would include domestic use of the water as considered by Miranger and Gladwell (1986), Maskowitz et al. (1986) or Taylor et al. (1986) and wildlife effects as discussed elsewhere in report. However, the relationships between the surface water chemistry and fisheries biota are at a higher level of definition (see Kelso et al. 1986) and the water chemistry criteria may actually be quite similar for relationships to other areas of concern. This section, therefore, will depend heavily upon the present knowledge or models of the interactions between acidic deposition, water chemistry and fisheries responses as reviewed in earlier sections of this report.

As shown in Tables 4, 5, 6 and 7, biological community structures differ as habitat acidity increases below pH=6. In the extensive surveys reported by Kelso et al. 1986, the numbers of fish species also decline most significantly below pH 6. It is thus not possible to define a single pH level that can be employed as a specific criteria where an aquatic resource can be considered to be at total risk. On a regional basis, it is more meaningful to define what portion of the aquatic ecosystem would be expected to attain degrees of acidification based on system sensitivity and acidification stress. The biosystem consequences of the acidification can then be inferred in terms of levels of losses.

Kelso et al. (1986) have estimated that there are more than 700,000 lakes in Canada east of the Ontario-Manitoba border and south of 52°N. This region receives greater than background levels of sulphate deposition that have been shown (previous sections this report) to correlate most significantly with respect to surface water alkalinity and, therefore, acidity. Existing water chemistry survey data have been employed to extrapolote to obtain an estimate of the profile of lake, or surface water sensitivity.

Marmorek et al. (1985) has extended the models of Henrikson (1979), Thompson (1982), and Wright (1983) to employ the present observation of cation concentrations, sulphate concentrations, and sulphate loading to produce a simulation of expected steady state alkalinity and therefore, acidity of surface waters. The accuracy and limitations of this model have been reviewed by Minns and Kelso (in press) and a scenario of simulations have been carried out using reasonable estimates of extremes of the dependent parameters. A major uncertainty in this simulation is the value of the F factor, i.e. the cation weathering. An additional, and in long-term (decade or longer) more important uncertainty, is the change or rate of change of the F-factor. If cation depletion occurs, the F-factor would decrease leading to more rapid rates of acidification under a given acidic stress.

Taking these factors into consideration, Minns and Kelso (in press) have produced extreme estimates of the portions of the lakes of eastern Canada that would be expected to be acidified, i.e. eventually attain a pH < 5, under a range of sulphate loadings. Because existing evidence indicates that sulphate retention in sensitive watersheds of the Canadian Sheild is not significant, the authors expect that a more realistic estimate lies towards the upper bounds of this simulation.

Based on the extrapolation of limited surface water surveys to the total lake inventory, approximately 14,000 lakes are presently estimated to be acidified (pH$<$ 5). Because of the limited surveys, confidence limits cannot presently be assigned to this estimate. The simulation indicates that if present sulphate loadings having a peak

value of about 36 kg SO_4/ha.yr (wet plus dry) are continued, 10,000 to 40,000 additional lakes will eventually become acidic. The presently proposed "target loading" of 20 kg SO_4/ha.yr (wet only) of deposition would be expected to result in a worst case additional loss of up to 15,000 lakes in the future. A reduction of the peak deposition rates to about 12 kg SO_4/ha.yr would be needed to assure no further losses of highly sensitive lakes to acidification and to assure a recovery of presently acidified, less sensitive lakes.

If the simulated lake acidification losses are considered at losses to angling opportunities, Minns and Kelso (in press) conclude that the present sulphate deposition rate will eventually lead to a loss of between 1.8 and 4.5 million angler-days in the area of eastern Canada. They have converted the loss into a potential loss of expenditures related to these anging-days of between \$59 and \$144 million.

These estimates, based on model simulations of resource-at-risk, must be considered to be conservative. They are based on a simulation that does not consider a possible depletion of acid neutralizing capacity of watersheds as a result of acidic deposition. Arp 1983, and Booty and Kramer 1984 suggest that there is likely to be a fixed reservoir of basic material that may be depleted. If this process does occur, the losses of lakes to acidification would eventually be greater than predicted by the simulation. Furthermore, much work needs to be done to assemble a more accurate inventory of lakes, at least for eastern Canada.

Because inventories of lake resources are not available on regional or drainage basin scales, lake surveys to predict assessments of the resources at risk, such as that reported by Kelso et al. (1986), must contain an unknown uncertainty. While this lack of an adequate basic inventory may account for the variances between the estimates of Kelso et al. (1986) and by Fraser (in press), such estimates do serve to indicate the large magnitude of the freshwater resource that is at risk in eastern Canada.

3.8 Need for Further Information

Throughout the development of this Issues Assessment Report, it became evident that, while a large amount of new research results have become available since the writing of the MOI report, there remain several areas where information continues to be insufficient to support definitive conclusions on cause-effect relationships. It is not the purpose here to outline the needs for further research in detail but rather to point out the broad general areas where information is required. Because the areas are generally interdependent, it is not possible to assign specific priority.

A. The evidence of changes or trends in aquatic chemistry in response to LRTAP continues to be lacking in confidence due to serious deficiency in both the water chemistry monitoring and the atmospheric deposition measurements. Acidification models require further development with more attention being given to prediction in the long term (decades or greater), but the ability to validate these models will depend critically on the existence of a highly reliable time series data base and a better understanding of the spatial variabilities in the data.

B. Factors that remain uncertain for both the direct evaluation of acidification effects and the acidification simulation models are the relative influence of within-system, alkalinity generation and variations in the rate of weathering of basic materials. Reactions including sulphate reduction or adsorption, nitrogen assimilation and basic cation exchange with lacustrine sediments are important. Moreover, the variations of these factors in relation to the deposition of acidifying substances is not clearly understood. These reactions must be defined for the long term for both groundwaters and surface waters as well as for the short term for surface waters.

C. The biological consequences of episodic events of increased acidity, increased concentration of free metals and reduced ionic concentrations must be established. The critical periods in relation to life stages and frequency of recurrence must be identified. These periods need to be quantified for all forms of biota, not just for fish.

D. The food chain responses to acidification or acidification related processes of metal or organic toxicity must be more precisely defined. Analyses of these responses must be extended into the aquatic-terrestrial species of amphibians, waterfowl and mammals. Critical levels of acidity and/or concentrations of toxic materials should be established that can be related to measured water chemistry variables.

E. The specific understanding of the responses of organisms to the acidification stress must be translated into a quantitative definition of regional or larger scale impacts on population viabilities. These translations must consider the food chain relationships of item D.

F. The biosystem responses to a reversal of acidification due to emission control or other mitigation management must be defined. Selective species responses should be understood for proper management of the resource.

G. A more exact inventory of the aquatic resources of Canada is required so the resources-at-risk in relation to acidification stress can be better defined. This inventory should include the physical water resource and associated biological community components.

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